Dynamic transition in etching with poisoning

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We study a lattice model for etching of a crystalline solid including the deposition of a poisoning species. The model considers normal and lateral erosion of the columns of the solid by a flux of etching particles and the blocking effects of impurities formed at the surface. As the probability p of formation of this poisoning species increases, the etching rate decreases and a continuous transition to a pinned phase is observed. The transition is in the directed percolation (DP) class, with the fraction of the exposed columns as the order parameter. This interpretation is consistent with a mapping of the interface problem in $d+1$ dimensions onto a d-dimensional contact process, and is confirmed by numerical results in $d=1$ and $d=2$. In the etching phase, the interface width scales with Kardar-Parisi-Zhang (KPZ) exponents, and shows a crossover from the critical DP behavior ($W\sim t$) to KPZ near the critical point, at etching times of the order of $(p_c-p)^{-\nu_\parallel}$. Anomalous roughening is observed at criticality, with the roughness exponent related to DP exponents as $\alpha_c=\nu_\parallel/\nu_\perp>1$. The main differences from previously studied DP transitions in growth models and isotropic percolation transitions in etching models are discussed. Investigations in real systems are suggested.

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I. INTRODUCTION

The study of interface growth has attracted increasing interest in the last years due to the potential technological applications and to the fundamental role in nonequilibrium statistical physics [1–4]. One of the important problems in this field is the dynamics of solid surfaces in contact with an etching solution, which motivated various works [5–11], usually considering discrete atomistic models with the basic mechanisms of the real processes represented by local etching rates, accounting for the effects of binding energies, anisotropy, concentration of the solution, etc. In the continuum limit, the morphology of the resulting solid-liquid interface may be described by one of the existing theories for interface growth [2–4].

In some cases, the etching of a solid is accompanied by the precipitation of an inert product on its surface, which may form clusters that act as a mask for the corrosion process. This is expected, for instance, in the etching of Si by NaOH, as discussed in Refs. [12,13]. The presence of blocked sites was also considered in a model for a passive layer formation between solid lithium and an acid solution [14–16]. The growth followed stochastic rules similar to those of the Eden model [2,4], the blocking species being produced at the interface. As the rate of formation of that species was increased, a transition to a blocked phase was observed. That transition was in the isotropic percolation class. On the other hand, Sapoval and co-workers considered the etching of a disordered solid by a solution of finite volume [6–8], in which the atoms exhibit random strengths against corrosion. Connections to isotropic percolation [17] and gradient percolation [18] processes were shown theoretically [19].

Anisotropic fluxes of etching particles are also important in many real processes. For instance, a recent work of Hwang and Redner have considered a bias in the diffusive motion of an acid during the dissolution of a solid [9]. Such a bias may represent the effect of an electric field on ionized particles, a gravitational field or pressure gradient on a flowing fluid. In a simplified way, these mechanisms are also represented in the model of Mello *et al.*, which considers a vertical flux of etching agents towards the surface of a crystalline solid and the corrosion of highly exposed regions in the neighborhood of the collision points [11].

This scenario motivates the present study of the effects of deposition of a poisoning species during the etching of a solid surface by a perpendicular flux of the etching particles. We will extend the model for etching of a crystalline solid of Ref. [11], allowing for the formation of a poisoning species which locally blocks the erosion. The presence of lateral growth indicates that the model is in the Kardar-Parisi-Zhang (KPZ) universality class of interface growth [20], which was confirmed numerically [11]. Here, the reactions between the solute and the solid may lead to the formation of the chemical species that blocks the etched column. This species may be removed only as a consequence of future etching processes at its neighborhood.

The model presents a continuous transition between an etching regime and a regime with blocked surfaces. We will study this transition numerically in two- and three-dimensional solids, showing that it belongs to the directed percolation (DP) universality class [22–25]. This is confirmed by the mapping of the etching process of a $(d+1)$-dimensional solid onto a d-dimensional contact process (CP). It contrasts with the isotropic percolation transition observed in the models for passive layer formation over lithium films by Badiali and co-workers [14–16], although those systems were also in the KPZ class in the growth regimes. As will be discussed below, there are also many differences between the DP transition of the present model and directed percolation depinning of interfaces growing in quenched disordered media [26,27]. One particularly important difference is that the present model also shows DP transitions in $2+1$ dimensions.

From the theoretical point of view, this work extends the discussion of a new type of pinning transition related to DP,

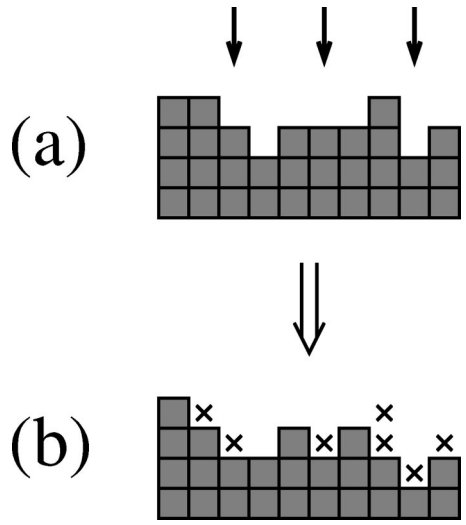

FIG. 1. (a) Examples of etching attempts of the original model (Ref. [11]) in $d=1$, in which the selected columns are indicated by arrows. (b) The solid after normal erosion at the top of the selected columns and lateral erosion of higher neighboring columns. The positions of removed atoms are indicated by crosses.

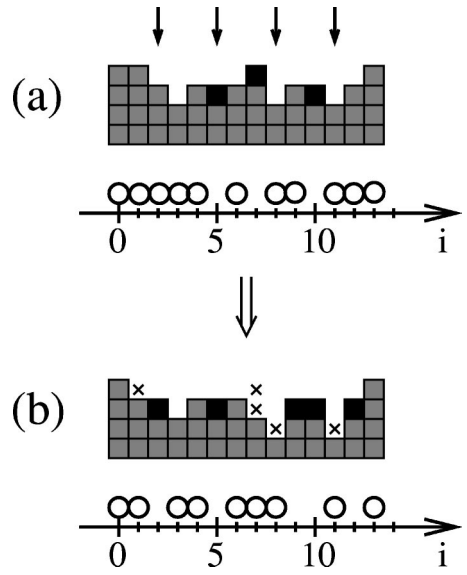

FIG. 2. (a) Examples of etching attempts of our model in $d=1$, with selected columns indicated by arrows, A particles in gray and B particles in black. The corresponding configuration of the associated one-dimensional particle-hole problem is shown below the solid, with particles (circles) corresponding to top A and holes corresponding to top B. (b) The solid after the etching at the selected columns, with positions of removed atoms indicated by crosses. Normal erosion at column 2 and lateral erosion at columns 9 and 12 left B particles at those columns. In column 7, a B particle was removed by lateral erosion. The configuration of the associated particle-hole problem is also shown.

introduced in a recent study of a ballisticlike deposition model with an inactive species [21]. The observation of critical anomalous roughening extends the conclusions of that work concerning interface width scaling. Although our model was not proposed to represent any particular experimental process, it shows some differences between the pure system's and the critical system's behavior which may be investigated experimentally. One possibility is the critical anomalous roughening and the consequent time dependence of the local interface width. In view of the absence of clear experimental realizations of DP transitions, we believe that the present work will motivate investigations in etching processes and related problems. To our knowledge, this is the first work that suggests a DP transition in etching processes.

The rest of this work is organized as follows. In Sec. II, we will present our model, discuss the related theory, the mapping onto a contact process, and the relations with other roughening transitions in growth models. In Sec. III, we will present numerical results for a two-dimensional solid, including the discussion on the critical anomalous roughening. Although this case is not realistic, it is much better for numerical studies, providing accurate estimates of critical exponents and giving support to our interpretation of the transition. In Sec. IV, we will present some results for a three-dimensional solid. In Sec. V, we will summarize our results and present our conclusions.

II. MODEL AND RELATED THEORY

The original model for etching of a crystalline solid [11] is illustrated in Fig. 1. The solid is assumed to have a square lattice structure in the $(1+1)$-dimensional version of the model and a simple cubic structure in $2+1$ dimensions. At each etching attempt, a column i of the solid, with current height $h(i)\equiv h_0$, is randomly chosen. Then its height $h(i)$ is decreased by one unit (supposing etching in the downward direction): $h(i)\rightarrow h_0-1$. This process will be called normal or vertical erosion. Finally, any neighboring column whose height is larger than h_0 is eroded until its height becomes h_0. This process will be called lateral erosion and represents the etching of regions of the interface which were exposed before the normal erosion process. Notice that lateral erosion is always induced by normal erosion.

The rules of the extended model studied in the present work are illustrated in Fig. 2 for a two-dimensional solid. This solid contains a single chemical species A, similarly to the original model, and any column with a top A is said to be exposed. An erosion process (normal or lateral) at a certain column may leave an inactive particle B at the top of that column, with probability p. Any attempt of normal erosion at a column with a top B is rejected, i.e., species B blocks the interface for this process. However, a column with a top B will be subject to lateral erosion if there is a particle A with an empty neighbor below the top B. Thus, lateral erosion of a column with a top B occurs after normal erosion of a neighboring column with a top A and with smaller height. For instance, normal erosion at the column with $i=8$ in Fig. 2 leads to lateral erosion at the blocked column with $i=7$ and at the exposed column with $i=9$. In this model, one time unit corresponds to one attempt of normal erosion per column.

In the limit of large substrates and long times, the original etching model of a $(d+1)$-dimensional solid belongs to the

KPZ class of interface growth in $d+1$ dimensions [11]. This theory proposes the Langevin-type equation [20]

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h + \frac{\lambda}{2}(\nabla h)^2 + \eta(\vec{x},t), \tag{1}$$

known as KPZ equation, as a hydrodynamic description of kinetic surface roughening. In Eq. (1), h is the height at the position $\vec{x}$ in a d-dimensional substrate at time t, ν represents a surface tension, λ represents the excess velocity, and η is a Gaussian noise.

In discrete models, the interface width, which characterizes the roughness of the interface, is defined as

$$W(L,t) = \left[\left\langle \frac{1}{L^d} \sum_i (h_i - \overline{h})^2 \right\rangle \right]^{1/2} \tag{2}$$

for deposition (or erosion) on a d-dimensional substrate of length L (h_i is the height of column i at time t, the bar in $\overline{h}$ denotes a spatial average, and the angular brackets denote a configurational average). For short times it scales as

$$W \sim t^{\beta_K}, \tag{3}$$

and for long times, in the steady state regime, it saturates at

$$W_{sat} \sim L^{\alpha_K}. \tag{4}$$

In Eqs. (3) and (4), index K refers to KPZ theory, which represents our discrete models in the continuum limit (below the transition points). In $d=1$, the exact KPZ exponents are $\beta_K = 1/3$ and $\alpha_K = 1/2$ [2,20].

For sufficiently high values of the probability p and after a sufficiently large etching time, the interface gets completely blocked by B particles and, consequently, the etching process stops. This dynamic transition between an etching (or growth) phase and a blocked phase is indeed observed in our simulations (Secs. III and IV). Similar transition was recently found in a ballisticlike growth model with an inactive species [21]. In that case, the deposition model in a d-dimensional substrate could be mapped onto a CP in d dimensions, suggesting that the transition was in the universality class of DP [24,25].

It is possible to define an analogous mapping for the present model. First, any column of the solid with a top A is associated to a particle in a d-dimensional lattice, while a column with a top B is associated to a vacant site. This relation is also illustrated in Fig. 2. Normal and lateral erosion in the etching model give rise to particle annihilation and offspring production in this particle-hole system. The deposition of a particle B after normal erosion corresponds to the annihilation of a particle in the CP. On the other hand, normal erosion may be followed by lateral erosion of a neighboring column with a top B, which leaves a top A there (with probability $1-p$). It corresponds to offspring generation in the CP process. One difference between the CP associated to the ballisticlike deposition model [21] and to the present etching model is that, in the present case, the generation of two or more offspring is possible. This is the case when normal erosion is followed by lateral erosion of two or more blocked columns and leaves those columns exposed. This CP may also include particle diffusion, since normal erosion may form a top particle B while subsequent lateral erosion of blocked columns leave them exposed. Finally, notice that the blocked state, in which all columns have a top B, corresponds to the absorbing state of the CP.

It is not possible to obtain an analytical relation between the probability p and the rates of annihilation and offspring production in the CP because they depend on the height distributions of the interface problem. However, despite the particular features of this CP, it has no additional symmetry [24,25] that could suggest a universality class of the transition different from DP, as will be confirmed by our numerical results. This shows again the robustness of the DP class.

Here it is important to stress the differences between this transition and those of related growth models in the DP class [2,26–29] or in the isotropic percolation class [6–8,14–16].

First we notice that KPZ scaling in the growth regime is also observed in the models for passive layer growth over lithium films [14–16], since lateral growth is allowed. They were extensions of the Eden growth model, where any active particle of the cluster may grow in any direction with equal probability. At criticality, the blocking particles form an isotropic percolation cluster, which was confirmed numerically [14,15]. This illustrates the fact that KPZ scaling in the growth regime does not necessarily lead to DP transitions.

Another important type of transition is the so-called directed percolation depinning (DPD) [2,26,27], in which KPZ growth occurs in a disordered medium. The interface gets pinned if the impurity concentration exceeds the DP threshold, generating a blocking cluster perpendicular to the growth direction. The relation to DP is restricted to $1+1$ dimensions. At criticality, the growth is expected to be described by the quenched KPZ equation [28,29], in which the thermal noise in Eq. (1) is replaced by a quenched noise. Moreover, the active phase of DP corresponds to the pinned phase of the interface problem, while in the absorbing phase of the impurity system the growth continues indefinitely. Some models with competition between aggregation and desorption also show transitions in which the regime of film growth corresponds to the absorbing DP phase [30,31].

On the other hand, our model shows DP transitions in all spatial dimensions, with the pinned phase corresponding to the absorbing phase of DP and the etching phase corresponding to the active one. Consequently, very different critical behavior of quantities such as etching rate and interface width are observed. Many differences were already discussed in the work on the ballisticlike deposition model with a poisoning species [21]. Here we will also show that the critical roughness exponent [Eq. (4)] of our model is the reciprocal of the exponent in the DPD models (Sec. III).

III. NUMERICAL RESULTS IN 1+1 DIMENSIONS

We simulated the two-species etching model in square lattices with a large lateral length $L = 2^{16}$ to avoid finite-size effects. Simulations in relatively small lattices ($L \leqslant 2048$), until the steady state regions, were performed for $p = 0.15$

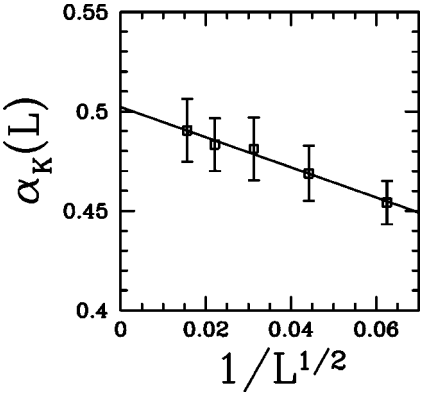

FIG. 3. Effective exponents $\alpha_K(L)$ vs $1/L^{1/2}$ for the $(1+1)$-dimensional model with $p=0.15$. The straight line is a least squares fit of the data.

and at the critical point. The etching rates, the fractions of exposed columns, and the interface widths were typically averaged over 10^3 different realizations (10^4 realizations in small lattices) with $p=0.15$.

First we consider the model at the etching phase with relatively small p. In order to estimate exponent α_K in Eq. (4) and confirm KPZ scaling, we defined the effective exponents

$$\alpha_K(L)=\frac{\ln[W_\infty(L)/W_\infty(L/2)]}{\ln 2}. \tag{5}$$

In Fig. 3 we show $\alpha_K(L)$ versus $1/L^{1/2}$, which gives $\alpha_K=0.5$ (the KPZ exact value) in the limit $L\to\infty$. The $1/L^{1/2}$ correction was chosen among other correction terms in the form $L^{-\Delta}$, with $\Delta>0$, and was the one that provided the best linear fit of the data. This extrapolation procedure was previously applied to the study of ballistic deposition [32]. KPZ scaling was also obtained in the study of the ballistic-like deposition model with an inactive species [21], but with stronger corrections to scaling. It is important to mention that our results contrast with the p-dependent exponents found in previous works on related two-species models with inactive species [33].

When p increases, the etching rate decreases due to the increase in the density of particles B at the surface. This rate decays as the fraction ρ of the surface exposed for etching, i.e., the fraction of top A, which is the order parameter of the etching-blocking transition. ρ is shown in Fig. 4(a) as a function of p and is expected to obey the scaling form

$$\rho\sim\epsilon^{\beta}, \quad \epsilon\equiv p_c-p. \tag{6}$$

In order to fit our data to Eq. (6), we calculated a series of effective exponents

$$\beta(p,p')\equiv\frac{\ln[\rho(p)/\rho(p')]}{\ln(\epsilon/\epsilon')}. \tag{7}$$

It is expected that, with the correct choice of p_c, $\beta(p,p')$ will converge to the asymptotic exponent β. In Fig. 4(b) we show $\beta(p,p')$ versus p_c-p, with $p_c=0.303\,18$, considering

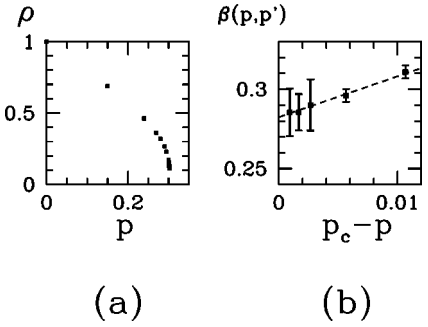

FIG. 4. (a) Fraction of exposed columns of the solid vs probability p in one-dimensional substrates. (b) The corresponding effective exponents $\beta(p,p')$ near the critical point, with $p_c=0.303\,18$. The dashed line is a least squares fit of the data.

data in the range $0.28\leqslant p\leqslant 0.3025$. Linear fits of $\beta(p,p')$ versus ϵ for other values of p_c were used to calculate the error bars of p_c and β. We obtained the estimates $p_c=0.303\,18\pm0.000\,05$ and $\beta=0.283\pm0.010$ under the condition of a linear correlation coefficient larger than 0.999 in those fits. It confirms that the transition is in DP class. For comparison, the best known estimate of exponent β for DP is $\beta=0.276\,486\pm0.000\,008$ [34].

In the etching phase ($p<p_c$), as the transition point is approached, the interface width scaling shows DP and KPZ features in two characteristic time intervals. At short times, W rapidly grows due to the increasing fraction of blocked columns, which give rise to increasingly large height differences. This is typical of critical DP because the fraction of exposed columns decreases slowly (algebraically). After a characteristic time of the order of the longitudinal correlation length $\epsilon^{-\nu_\parallel}$, KPZ scaling is expected. Notice that the etching direction corresponds to the longitudinal direction of the CP (or DP) problem, while the directions parallel to the interface correspond to the transversal directions of the CP.

This crossover is illustrated in the plot of Fig. 5(a), in which W is shown as a function of the scaling variable $x\equiv\epsilon^{\nu_\parallel}t$, with $\nu_\parallel=1.733\,847$ [34]. In Fig. 5(b), we show the linear growth of W at criticality ($p_c=0.303\,18$). Before the crossover in Fig. 1(a), the growth of W is also approximately linear, while for larger times it follows Eq. (3) with $\beta_K\sim1/3$.

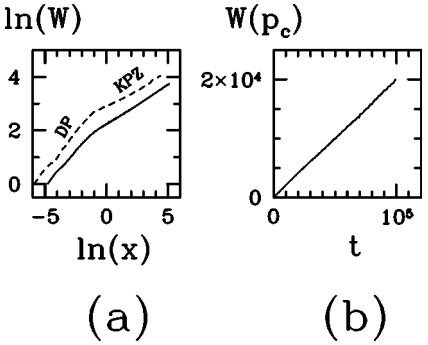

FIG. 5. (a) $\ln W$ vs $\ln x$, with scaled time $x\equiv t\epsilon^{\nu_\parallel}$, in $d=1$, for $p=0.24$ (solid line) and $p=0.27$ (dashed line) ($L=65\,536$). (b) Interface width W vs time t at the critical point in $d=1$, with $p_c=0.303\,18$.

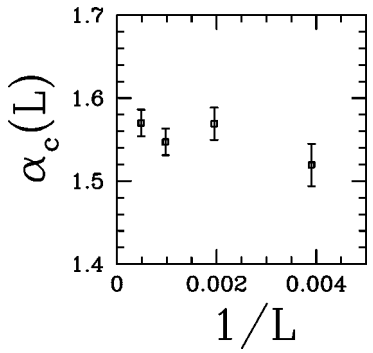

FIG. 6. Effective exponents $\alpha_c(L)$ vs $1/L$ for the $(1+1)$-dimensional model at criticality ($p_c=0.303\,18$).

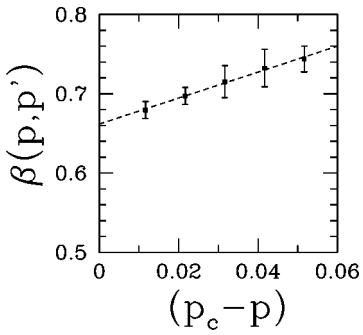

FIG. 7. Effective exponents $\beta(p,p')$ near the critical point in $d=2$, for $p_c=0.621\,65$. The dashed line is a least squares fit of the data.

In a study of the dynamical phase transitions in the Domany-Kinzel cellular automata, de Sales *et al.* [35] considered a related problem. They calculated the transition points with accuracy by mapping cellular automata patterns onto an interface problem, which shows a crossover from random deposition behavior ($\beta=1/2$) to critical DP behavior ($\beta=1$).

At the critical point in finite lattices, a finite fraction of the samples are etched indefinitely, attaining the steady state regime. At certain probability $p<p_c$, the transversal correlation length $\xi_\perp\sim\epsilon^{-\nu_\perp}$ equals the lattice length L and saturates at this value in the whole critical region, as expected from finite-size scaling theory [36,37]. On the other hand, height fluctuations are of the order of the longitudinal correlation length, then $W\sim\epsilon^{-\nu_\parallel}$. Thus, the saturation widths [Eq. (4)] at the critical point scale with a roughness exponent

$$\alpha_c=\nu_\parallel/\nu_\perp. \tag{8}$$

In order to confirm this relation numerically, we calculated effective exponents $\alpha_c(L)$ as in Eq. (5). These exponents are shown in Fig. 6 as a function of $1/L$, with $256\leqslant L\leqslant 2048$. They suggest $\alpha_c=1.57\pm0.03$ as $L\to\infty$, in agreement with the value obtained from estimates of DP exponents, $\nu_\parallel/\nu_\perp\approx1.581$ [34].

Since $\nu_\parallel>\nu_\perp$ in any dimension, $\alpha_c>1$, which suggests that the critical system has anomalous surface roughening (see Ref. [38], and references therein). However, at the critical point, the fraction of surviving deposits in large lattices is very small and the width distributions are very broad. Thus, much longer simulation times would be necessary to estimate the local roughness exponent of this system with reasonable accuracy. On the other hand, important qualitative features of the local interface width near p_c are observed, but their discussion will be postponed to Sec. IV because the $(2+1)$-dimensional system is more interesting for real systems' applications.

Note that the above value of α_c is the reciprocal of the critical roughness exponent of models which describe directed percolation depinning in $(1+1)$-dimensional disordered media [26,27]. This is expected because, in that case, the DP critical cluster runs in the direction parallel to the surface, while in our model it runs along the growth direction, which is perpendicular to the surface. The DPD models do not show anomalous roughening.

For completeness, we mention that results in the blocked phase ($p>p_c$) confirmed the DP class of the transition. In this regime, the etching process fails when the whole interface is covered with *B*. The average depth after complete poisoning attains a limiting value H_s which is of the order of the absorption time in the CP. Thus it scales as

$$H_s\sim(-\epsilon)^{-\nu_\parallel}. \tag{9}$$

Our numerical estimate $\nu_\parallel=1.75\pm0.02$, obtained with the above estimate of p_c, is also consistent with DP within error bars [34]. The interface width of the blocked interfaces also scales as Eq. (9).

IV. NUMERICAL RESULTS IN 2+1 DIMENSIONS

We simulated the two-species etching model in simple cubic lattices with lateral lengths ranging from $L=256$ to $L=1024$. Average quantities were typically measured over 100 different realizations.

The fraction ρ of the surface exposed for etching (top *A*) also decreases with p until the critical point at $p_c=0.621\,65$. In Fig. 7, we show the effective exponents $\beta(p,p')$ [Eq. (6)] versus ϵ, using this value of p_c. The asymptotic estimate is $\beta\approx0.66$, which is 13% above the best known DP estimate $\beta=0.584\pm0.004$ [39]. It may be possible to improve this estimate by calculating accurate densities ρ nearer p_c and by considering more general forms of scaling corrections, instead of the above linear correction in ϵ. However, much longer simulations in very large lattices would be necessary.

The interface width shows the same scaling properties observed in $d=1$: a crossover between DP and KPZ behaviors at $t\sim\epsilon^{-\nu_\parallel}$ below the critical point, and DP behavior (linear increase of W) at p_c.

Another interesting feature of this system is the time dependence of the local interface width w near criticality. In simulations on lattices with $L=2048$, w was averaged over finite-size boxes of lateral lengths $2\leqslant l\leqslant512$ spanning the whole surface. In Fig. 8 we show w versus l for the original etching model ($p=0$) at $t=200$ and $t=2000$. It illustrates the typical behavior of that quantity: a growth re-

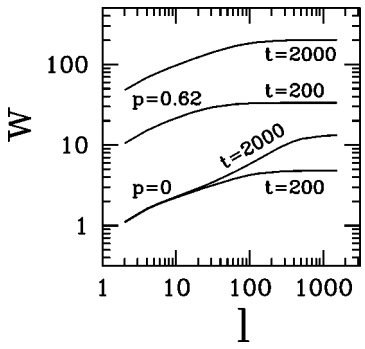

FIG. 8. Local interface width w as a function of box length l measured in lattices with total lengths $L=2048$, for $p=0$ (no impurities) and $p=0.62$ (near criticality), at two different times.

gion with $w \sim l^{\alpha_K}$ for small l and a crossover to a saturation value, which is the global interface width W at time t. Notice that, at small length scales, the local width w is nearly the same at both times, i.e., the height fluctuations have attained approximately constant values, characteristic of the dynamical process. The time evolution of w is restricted to large length scales. This is expected because the relaxation time of the structure factor increases with the wavelength λ, in the form λ^z [2,3].

We also show in Fig. 8 results for $p=0.62$, which is very near the critical point, also at $t=200$ and $t=2000$. Here we note two remarkable features: the values of w are much higher, indicating large height fluctuations even at narrow observation windows, and the values at small length scales still increase in time. It is explained by the decreasing number of exposed columns of the solid near criticality and reflects the anomalous roughening in this system.

The relevance of this qualitative analysis is that the local width is frequently measured in growth or etching experiments. If the above time dependence of w were observed in systems with one or more blocking species, they would suggest anomalous scaling and investigations of a possible dynamic transition.

V. CONCLUSION

We studied a model for etching of a solid (particles A) with formation of a blocking species B as a consequence of reactions in a solid-liquid interface. The etching rate decreases when the probability of creation of B particles increases and is zero after a certain critical probability p_c. This transition is continuous and is in the DP universality class. In the etching phase, the interface width has an approximately linear growth at small times and crosses over to KPZ scaling after a characteristic time of the order of the longitudinal correlation length of an associated contact process. The longitudinal correlation exponent $\nu_\parallel$ governs the divergence of the height of blocked deposits above p_c. Anomalous roughening is observed at criticality, with a global roughness exponent $\alpha_c > 1$ related to DP exponents [Eq. (8)]. Thus, near p_c, the local widths at small length scales increase in time, contrary to their behavior in the systems with normal roughening.

The DP transition in this model contrasts with the previously studied isotropic percolation transitions in etching models of disordered solids [6–8,19]. The comparison with growth models with production of poisoning species and isotropic percolation transitions [14–16] show that KPZ scaling in the growth regime is not a sufficient condition for the DP critical behavior. The transition in the present model also has remarkable differences from the depinning transitions of growing interfaces in disordered media [2,25–27] and in some models with aggregation and desorption [30,31]. Here, the etching (pinned) phase parallels the active (absorbing) phase of DP, while the opposite relation was found in those systems. Previously, a DP transition similar to the present one was observed in a ballisticlike deposition model with a poisoning species [21], for which a correspondence to a contact process was also proposed. Once again, it illustrates the robustness of the DP class [25,40]. For applications to real systems, this robustness suggests that DP transitions may be observed in etching processes with formation of blocking species even if the etching mechanisms are not represented by our particular model, but with anisotropic flux of the etching agents.

ACKNOWLEDGMENTS

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